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Assessing PET chemical recycling via neutral hydrolysis: A techno-economic and GHG emissions study

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ABSTRACT

Chemical recycling of PET waste is a promising approach for the recovery of high-quality monomers that aims at improving the circularity of plastic production. Solvolysis technologies are being developed and include hydrolysis with acid/base catalysts or enzymes to promote depolymerization reactions into PET constituent monomers, terephthalic acid (TPA) and ethylene glycol (EG). However, the use of electricity, steam and organic solvents/chemicals has been identified as main factor affecting the economic and environmental performance of these technologies. This paper studies an alternative neutral hydrothermal process (nHTP), which involves hydrolysis at high temperatures and pressures without the use of catalysts, looking into the impact of water recycling and EG recovery in terms of costs and GHG emissions. The results reveal that recovering EG pays off the higher CAPEX and OPEX and yields a significant reduction in wastewater treatment costs, resulting in a TPA production cost of 1–1.4 EUR/kg and a carbon footprint of 1–1.7 kg CO_2e/kg TPA. Although the process demands less electricity and steam compared to other hydrolysis technologies, fuel requirements for high temperature heat might limit its potential in the absence of sustainable heat options. Future studies are recommended to assess the utilization of lower-cost feedstocks (mixed waste and textiles), valorization of the solid by-product, and system integration possibilities to identify synergies between technologies in a broader chemical recycling context.

1. Introduction

Plastics are integral to modern society, enabling advances in healthcare, transportation, packaging, and technology through their versatility and low cost. Yet, inefficient waste management has led to widespread plastic pollution, while global plastic production remains largely fossil-based, with only about 10% derived from circular sources [1]. Of the 430 Mt. of plastics produced globally in 2024, polyethylene terephthalate (PET) accounted for 6% [1], in addition to roughly 78 Mt. of polyester manufactured for the textile sector [2]. This makes PET one of the major commodity polymers, widely used in packaging and fibers, and therefore a critical target for new recycling technologies aimed at reducing the environmental footprint of plastics. Mechanical recycling is suitable for clean and homogeneous PET waste streams, such as bottles recovered through deposit-return systems, but PET from mixed plastics and textiles requires chemical routes to recover high-quality materials [3].

PET ester linkages are readily cleaved through well-established solvolysis processes such as glycolysis, alkaline/enzymatic hydrolysis, and methanolysis, enabling efficient monomer recovery for virgin-grade PET production [4]. Several process modeling and techno-economic (TEA) and life-cycle (LCA) assessments have been conducted for these main PET solvolysis routes, indicating that although all three depolymerization processes can achieve similar monomer recoveries, glycolysis exhibits better economic and environmental performance due to a relative lower use of energy and chemicals, while methanolysis and hydrolysis have potential to provide higher monomer qualities [5–7]. Among the hydrolysis routes, which avoid the intensive use of organic solvents, a less explored route is neutral hydrolysis—also known as neutral hydrothermal processing (nHTP)—which employs only hot-compressed water (190–400 °C, 10–350 bar) as both reactant and reaction medium [8]. This route eliminates the need for added chemical agents while producing terephthalic acid (TPA) and ethylene glycol (EG), drop-in monomers for most existing polymerization units. Despite the

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potential advantages of nHTP, its benefits have not been systematically quantified, largely due to the lack of relevant experimental and process data. This gap makes it challenging to benchmark nHTP against other depolymerization pathways in terms of energy and material efficiency, and consequently to assess its economic viability.

Recently, detailed process design and continuous experimental results for nHTP have been reported in literature [9], providing valuable data for developing more reliable process models. Since this route does not employ catalysts to enhance depolymerization, water is required in excess to shift equilibrium towards the monomers, which makes wastewater management crucial to reduce freshwater consumption and improve the process performance. In a recent publication by our group [10], recirculation of the aqueous phase has been experimentally investigated as a strategy to reduce water consumption, enhance EG recovery, and potentially improve heat integration within the nHTP process, demonstrating that up to 67% recirculation is feasible without affecting the chemistry of the process. Still, there are no published studies to date that delve into nHTP process modeling and provide insights into process design, heat integration, and alternative configurations, factors that significantly influence TEA and LCA results.

In this study, we build upon these advances to provide a first-of-a-kind assessment of the techno-economic feasibility and carbon intensity of the nHTP technology for PET chemical recycling. For this, we use Aspen Plus modeling, published experimental data and inventory data to delve into the nHTP process design, estimate mass and energy balances, TPA production costs and carbon footprint of the process under different configurations. The results are discussed and compared with competing PET chemical recycling routes studied in literature, aiming at identifying similarities, limitations, and areas for future research that contribute to a more competitive technology basis in the chemical recycling landscape.

2. Methods

2.1. Process design

The nHTP process considered in this study is based on the continuous nHTP lab scale unit validated by Azuara et al. [9] and is modeled in Aspen Plus V12. *Ex ante* assessments of process interventions including scale-up considerations such as product separation, aqueous phase (AP) recirculation, heat integration and recovery of EG are conducted in Aspen Plus V12, as illustrated in Fig. 1.

The feed PET enters the process at 25 °C and 1 bar and is melted and pressurized to the reaction pressure using an extruder (EXT) and a gear pump (P2). The stream composes 100% PET and is modeled in Aspen Plus as a polymer using the POLYNRTL property package. The melted polymer mixes with hot, pressurized water in a mass ratio of 10:1 relative to the input PET. Since water is consumed in the depolymerization process and not all water is recirculated with the AP, additional make-up water is added by design specification so the water in the reactor feed is always equal to 10 times the mass of PET. The mixture is heated up in the furnace (F1) to be fed to the reactor (R1) operating at 310 °C and 165 bar. Decompression and cooling take place in three stages (V1, CR1, CR2) from which saturated vapor at different pressures is separated and used as heating medium in the AP pre-heaters (HX1, HX2, HX3) and mixed with the remaining aqueous separated from the TPA in the centrifuge (CN1). A fraction of the total AP is recirculated and the remaining (recirculation bleed) is sent to the EG recovery section. The solid TPA is then washed (SW) and dried (DR1) to meet purity specifications. In the EG recovery section three distillation units are included due to the low concentration of EG in the recirculation bleed (< 10 wt%). The mechanical vapor recompression unit (T1-MVR) is specified with a distillate to feed ratio of 0.85 to increase the AP concentration, and a crystallizer (CR3) and centrifuge (CN2) separate low-soluble by-products prior the second distillation unit (T2). T2 is specified to recover 99.9% of the water in the distillate, leaving a mixture of EG and remaining dissolved by-products as the bottoms product. Last, EG purification is carried out in the vacuum-distillation unit (T3) in

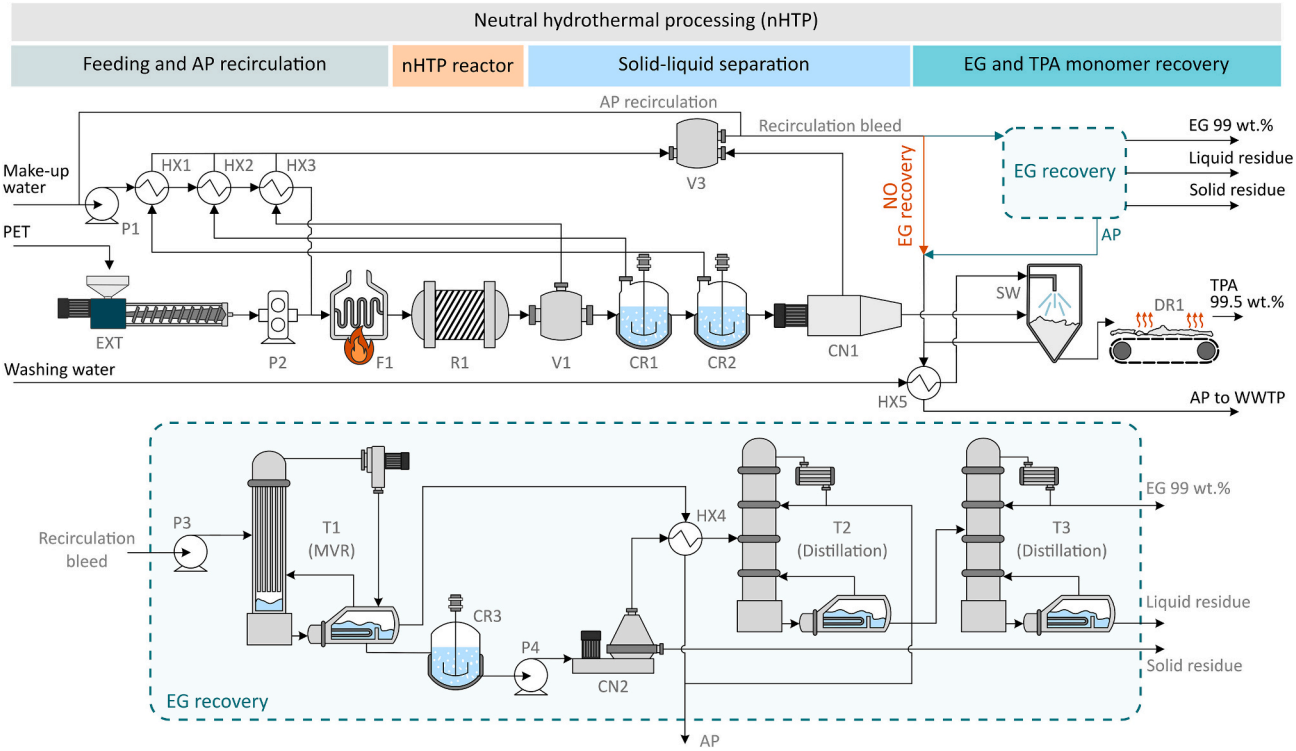

Fig. 1. Process flow diagram of neutral hydrothermal processing (nHTP) of PET.

which a target EG purity of 99 wt% is specified.

The simulated process is evaluated in terms of mass and energy balances by comparing two layouts:

- TPA & EG recovery: Full process including both TPA and EG recovery in which depolymerization by-products are separated from the aqueous phase as residues and the remaining wastewater is sent to WWTP.
- No EG recovery: EG is not recovered and only TPA is produced. The *Recirculation bleed* (Fig. 1) is sent to treatment at a wastewater treatment plant (WWTP).

The impact of AP recirculation in the overall mass and energy balances is evaluated up to 70% recirculation [10], having a fixed TPA purity of 99.5 wt% (polymer grade) and EG purity of 99 wt%. The main assumptions used are described as follows:

2.1.1. Plant size

The process is modeled at a scale of 24 tonnes per day (t/d) of PET feed, equivalent to 8 thousand tonnes per year (8 kt/y). This is in the range of small-scale plants considered in existing TEAs [6,11,12] that could fit individual plastic recycling sites in distributed configurations. For reference, the average production of waste PET in Europe from plastic recycling sites was estimated to 3.6 kt/y in 2022 [13]; while in the US, the estimate is around 11 kt/y from the largest materials recycling facilities [6]. A larger plant of 300 t/d (100 kt/y), representative of a more centralized configuration, is included in the study to evaluate the impact of the economy of scale.

2.1.2. nHTP reactor

The reactor is modeled based on experimentally reported molar balances obtained at 310 °C, 165 bar and a reaction time of 12 min, using pure PET and water as reactants [9]. It is assumed that additional secondary compounds entering the reactor from the recirculated AP do not influence the reaction yields and go through the reactor unchanged. The assumption is supported by Shabbir et al. [10] who observed no significant changes in the chemistry of the nHTP process upon AP recirculation up to 67%. In Aspen, the reactor is implemented as a user model connected to an Excel worksheet that contains the molar yields to calculate the reactor outputs. The molar yields from the experimental data in Table 1 (first column) must be adjusted since they result in a carbon excess of 4.2%, while hydrogen and oxygen balances are not reported since the water consumption is not measured. For this, the components with low yields that are not available in the Aspen Plus database (e.g./i.e. DTG, TGT and MHEI) are excluded, and the water consumption is estimated by minimizing the error in the hydrogen balance (solver tool in Excel). This procedure lowers the carbon imbalance to 2.4% due to the high molecular weight and carbon content of these compounds but leaves a remaining 0.2% excess mass in the overall balance. Since the mass balance must close to guarantee convergence, the error is distributed across all components (excluding water) proportionally to the yields to close the mismatch, resulting in the adjusted values in Table 1 which are the final input values for the Aspen Plus simulation. Once the composition of the reactor effluent is defined, enthalpy and other stream properties are calculated by Aspen Plus assuming isothermal operation and no pressure drop in the reactor.

2.1.3. Products separation and monomers recovery

The NRTL-Hyden-O'Connell (NRTL-HOC) property package is used as global method for its accuracy in predicting V-L equilibrium of mixtures containing organic acids, and a global chemistry is defined to include ionic dissociations (acetic and benzoic acid) and phase equilibria to predict precipitation of poorly soluble compounds. Regarding solid-liquid equilibrium, we use the data regression tool available in Aspen Plus to estimate binary NRTL parameters and equilibrium constant coefficients based on reported solubility measurements of the systems TPA/H_2O [14,15], TPA/EG [16,17], BHET/H_2O [18,19], BHET/EG [18] and IPA/H_2O [20] (Section S1, supplementary document). The parameter estimation is limited to binary systems since experimental solubility data of these compounds in mixtures of H_2O and EG is not available in literature. Furthermore, no experimental solubility data of MHET was found in literature, so in this case the solubility predicted by Aspen Plus could not be validated. More details regarding the process design and choice of process conditions can be consulted in the supplementary document, section S1.

Table 1
Molar yields of nHTP products at 310 °C and 165 bar.

Component	CAS Number	Molar yields from experimental data [a] [mol/100 mol PET input]	Molar yields adjusted [mol/100 mol PET input]
Water	7732-18-5	NR [b]	−181.772
Terephthalic acid (TPA)	100–21-0	94.231	92.309
Ethylene glycol (EG)	107–21-1	78.049	76.458
Acetaldehyde	75–07-0	9.282	9.093
Mono(2-hydroxyethyl) terephthalate (MHET)	1137-99-1	6.745	6.607
Diethylene glycol (DEG)	111–46-6	2.995	2.934
Isophthalic acid (IPA)	121–91-5	0.836	0.819
Benzoic acid	65–85-0	0.581	0.569
1,4-Dioxane	123–91-1	0.493	0.483
Bis(2-hydroxyethyl) terephthalate (BHET)	959–26-2	0.225	0.221
Ethanol	64–17-5	0.075	0.074
2-Butenal	123–73-9	0.018	0.018
Acetic acid	64–19-7	0.014	0.014
DGT (not available in Aspen Plus database)	–	1.100	–
TGT (not available in Aspen Plus database)	–	0.103	–
MHEI (not available in Aspen Plus database)	–	0.078	–
Error in elemental balance			
Carbon [%]	–	4.20	0.20
Hydrogen [%]	–	NR [b]	0.01
Oxygen [%]	–	NR [b]	0.01

[a] Based on Azuara et al. [9] estimated as the total molar flow reported for each component divided by the molar flow of PET used in the experiments (1 mol of PET is defined as 1 repeating unit).
[b] Not reported.

2.2. Economic evaluation

An economic evaluation is conducted to estimate the TPA production cost and assess the impact of the EG recovery process on the overall costs

Table 2
Summary of case studies in economic evaluation.

Case	Description	Scale [t/d PET feed]	AP recirculation	EG recovery
1	TPA & EG recovery -small	24	Yes	Yes
2	No EG recovery -small	24	Yes	No
3	TPA & EG recovery -large	300	Yes	Yes
4	No EG recovery -large	300	Yes	No

at different plant capacities. The cases considered are summarized in Table 2 and are evaluated in baseline, low and high-cost scenarios (Table 3).

The installed equipment costs are obtained from Aspen Economic Analyzer (APEA) in USD_{2019} and converted to EUR_{2024} using a conversion factor of 0.9 and the chemical engineering plant cost index ratio of the referenced years ($CEPCI_{2024}/CEPCI_{2019} = 1.31$). The reactor cost is not calculated in APEA but is estimated based on the reported base cost and scale factor for an HTL reactor [21], using a residence time of 12 min [9] to estimate the volume. The total capital investment is estimated by multiplying total installed equipment cost by a Lang factor of 4.64 (solid-liquid process). Regarding operating expenses (OPEX), the price ranges of feedstocks, utilities and products are listed in Table 3. The cost for wastewater treatment is calculated based on estimates for wastewater treatment plants (WWTP) in Denmark [22], where the fee is calculated as the sum of a base tariff and special contributions that must be paid when the discharged water exceeds certain limits of chemical oxygen demand (COD) as indicated in the following equation:

$$WWTP_{fee} = U_{base} + (COD - 1600)\frac{U_{COD}}{1000}$$

where $WWTP_{fee}$ is the fee in EUR/m^3; COD is in units of mg/L; and U_{base} and U_{COD} are the base tariff and the additional tariffs for COD in EUR/m^3 and EUR/kg respectively (factors for total nitrogen and phosphorous can be included but are not applicable in this case). For the high-cost scenario, a 20% increase is assumed since there is no better estimate available. Operation and maintenance (O&M) and other fixed costs (local taxes, insurance, plant overhead and administrative costs) are estimated based on typical methodology in literature [23]. The TPA production cost is then estimated using a net present value (NPV) analysis with a 10% discount rate, straight line depreciation and a plant lifetime of 25 years.

2.3. Carbon footprint estimation

Emissions from the process are estimated based on the results of mass and energy balances and the emission factors summarized in Table 4. Low and high emission factors are selected from the referred publications and values available in EcoInvent Database v3.11 using the IPCC 2021 Global Warming Potential over a 100-year time horizon (GWP100). The results are presented in kg CO_2eq/kg TPA produced and the impact of credits from the availability of by-products is discussed following a system expansion approach.

Table 3
Cost parameters for economic evaluation in baseline, low and high-cost scenarios.

Parameter	Baseline	Low cost	High cost
CAPEX	–	−10%	+10%
Feed PET sorted [EUR/t] [24]	575	520	630
EG credit [EUR/t] [25]	700	850	580
Process water [EUR/m^3] [26]	0.125	0.1	0.15
Cooling water [EUR/m^3] [26]	0.035	0.02	0.05
Electricity [EUR/MWh] [27]	60	50	100
Fuel (natural gas) [EUR/MWh] [28]	75	50	100
Efficiency furnace [%]	85	85	85
HP steam [EUR/MWh] [26]	45	30	60
LP steam [EUR/MWh] [26]	35	20	50
Waste disposal (landfill) [EUR/t] [29]	80	40	120
U_{base} (WWTP fee) [EUR/m^3] [22]	3.31	3.31	+20%
U_{COD} (WWTP fee) [EUR/kg] [22]	0.58	0.58	+20%
Operation hours per year [h]	8000	8000	8000
Operators per process section per shift	1	1	1
Number shifts	4	4	4
Process sections- TPA & EG recovery layout	5	5	5
Process sections- No EG recovery layout	4	4	4
Hourly labor costs EU [EUR/h] [30]	31.5	31.5	31.5
Hours per year per operator	2080	2080	2080

Table 4
Emission factors of material and energy flows used for carbon footprint estimation.

Carbon footprint	Low [kg CO_2e/unit]	High [kg CO_2e/unit]	unit
PET flake	0.686 [31]	0.908[a]	kg
Make-up water	2.00E-04[b]	0.0453 [31]	kg
Electricity	0.174[c]	0.187[d]	kWh
Steam	0.328[e]	0.398[f]	kWh
Natural gas	0.220[g]	0.254[g]	kWh
Cooling water [h]	1.632	3.222	MWh
Sodium hydroxide	0.903[i]	0.911[i]	kg
Sulfuric acid	0.091[i]	0.122[i]	kg
Sodium chloride	0.146[j]	0.266 [j]	kg
Wastewater treatment	0.750 [32]	1.250 [32]	m^3
Solid waste disposal	0.184[k]	0.455[k]	kg
TPA credits (virgin TPA)	2.17[l]	1.88 [31]	kg
EG credits (virgin EG)	1.94[m]	1.76 [31]	kg
Sodium sulfate credits	0.758[n]	0.528[n]	kg

[a] Ecoinvent database. Market for recycled PET flakes (RER).
[b] Ecoinvent database. Tap water Conventional Treatment, production (RER).
[c] Ecoinvent database. Electricity high voltage (DK).
[d] Average GHG emission intensity of electricity generation, EU level [33].
[e] Corresponds to 0.185 kg CO_2e/kg steam [31], converted to kWh basis assuming medium-pressure steam (2.03 MJ/kg).
[f] Ecoinvent database. Heat from Steam, Steam Production (RER).
[g] Ecoinvent database. Direct emissions of 0.202 kg CO_2e/kWh plus upstream emissions of 0.19 and 0.55 kgCO_2e/m^3 in DK and EU markets for the low and high respectively.
[h] Estimated based on assumed electricity consumption for cooling water pumping of 0.05 and 0.1 kWh/m^3 for the low and high scenarios respectively, a temperature approach of 5 °C, and the respective electricity carbon footprints in Table 4 [34].
[i] Ecoinvent database. Low production (RER) and high market (RER) for low and high values respectively.
[j] Ecoinvent database. Production (RER) and Market (RER) for low and high values respectively.
[k] Ecoinvent database. Waste PET landfill (GLO) and Solid waste incineration (DK) for low and high values respectively.
[l] Ecoinvent database. Purified TPA (RER).
[m] Ecoinvent database. EG (RER).
[n] Ecoinvent database. Market (RER) and Production (RER) for low and high values respectively.

With the aim of benchmarking, other PET chemical recycling technologies i.e. alkaline and enzymatic hydrolysis are included in the analysis using the mass and energy requirements reported in recent publications that assess their techno-economic and environmental feasibility [6,7,31]. These processes use PET flakes as feedstock, water as solvent and yield TPA as the main product, facilitating comparison with the nHTP process, but differ in the use of chemicals and the by-products generated. For a consistent comparison across technologies, only the reported material and energy requirements relative to the PET flake feedstock are used as input data to our analysis to compute the GHG emissions (see Table 9 in Supplementary document). Regarding the chemicals, the enzymatic hydrolysis process requires sodium hydroxide and sulfuric acid which are reported as net inputs [6,31], while for alkaline hydrolysis only make-up salt (sodium chloride) is reported because the process includes chemicals regeneration [7]. In terms of by-products, the enzymatic hydrolysis process generates sodium sulfate besides EG for which additional credits are considered for this technology.

3. Results

The next sections discuss the main results in terms of the impact of aqueous phase recirculation, overall mass and energy requirements, costs and carbon footprint of the process. For more detailed results of the Aspen Plus model, the reader is referred to the supplemental information (Sections S2 —S3).

3.1. Impact of aqueous phase recirculation in process performance

Fig. 2 presents the impact of aqueous phase recirculation in the overall mass balance, energy requirements and presence of contaminants for the process layouts with TPA and EG recovery and no EG recovery.

In terms of the overall mass balance (top figures), the comparison between the two layouts shows that the main difference is not only the EG product, but the solid and liquid residues separated as part of the EG recovery process. In the case without EG recovery (right figure), these components remain in the aqueous byproduct sent to wastewater treatment (AP to WWTP) representing an increase in mass between 3 and 10%. In both layouts, there is a significant reduction in make-up water and wastewater at increasing recirculation that highlights the clear benefit of this strategy in reducing the water footprint of the process. The yields of TPA and EG remain largely unchanged across AP recirculation ratios, which can be explained by the dominant effect of the low TPA solubility and the significant difference in volatility between water and EG. Although recirculation of the AP does not seem to affect the recovery of the monomers, it does have an impact on the yields of the solid and liquid residues being more favorable at high recirculation rates to enhance precipitation.

In terms of energy consumption (middle charts), both layouts have the same fuel and low-pressure (LP) steam requirements but differ in consumption of electricity, high-pressure (HP) steam and cooling water. The increase in fuel (Natural gas) consumption of the furnace with AP recirculation is explained by the increase in heat capacity of the reactor feed due to the accumulation of dissolved species upon recirculation, which occurs in both layouts independently of the EG recovery section. In the layout including EG recovery (left chart), electricity, HP steam and cooling water required to run the MVR and distillation units decrease at increasing AP recirculation due to the higher EG concentration in the recirculation bleed. Thus, when both monomers are recovered, high recirculation rates are clearly more favorable to

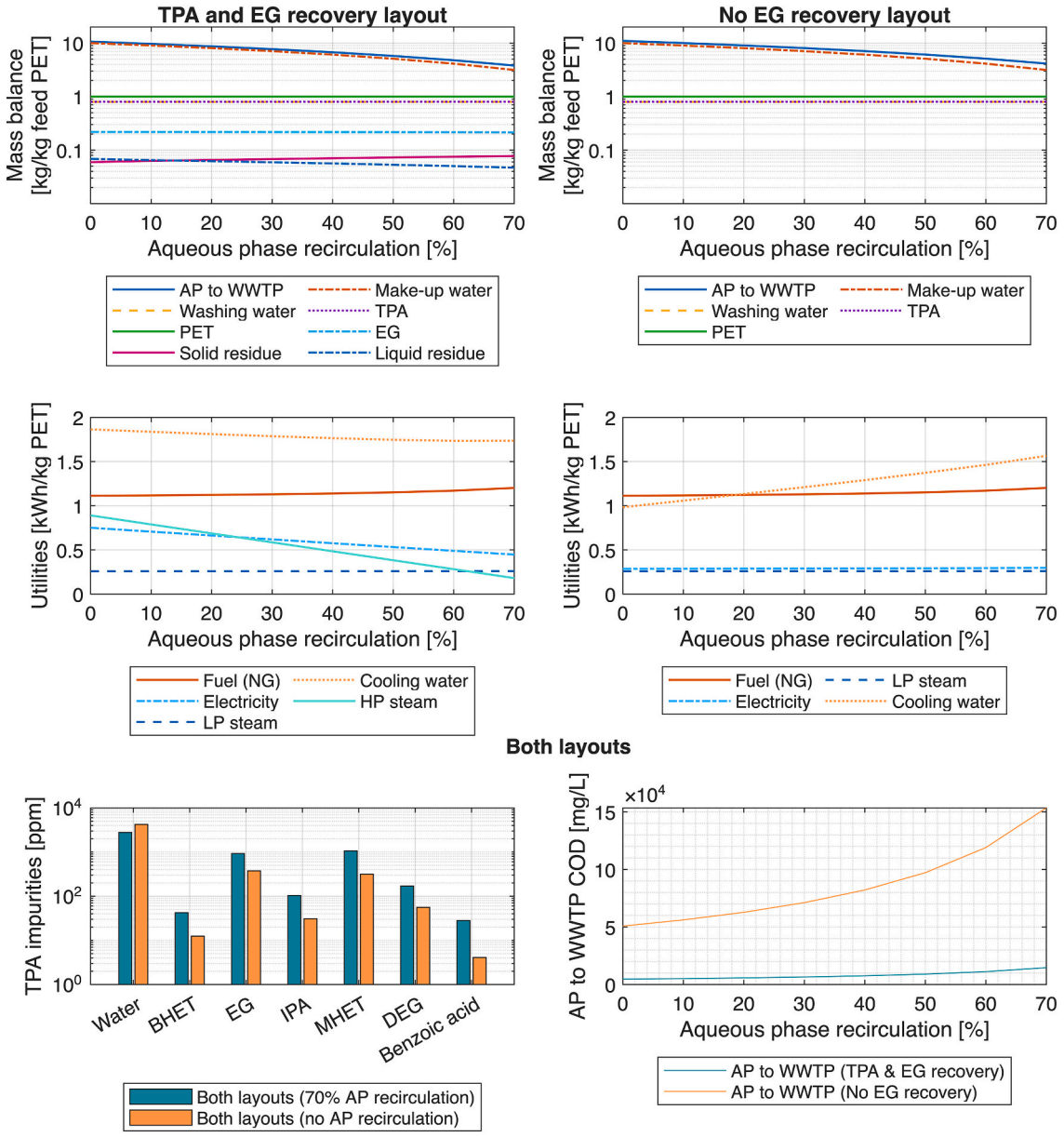

Fig. 2. Impact of aqueous phase recirculation on overall mass balance (top), energy requirements (middle) and TPA/AP contaminants (bottom) for the process with TPA and EG recovery (left) and with no EG recovery (right). The results at 70% AP recirculation are highlighted as these are used for the cost estimation and GHG analysis.

decrease energy consumption. For the layout in which only TPA is recovered (right chart), the increase in cooling requirement is related to the increase in the AP heat capacity for which lower recirculation rates would then be more favorable to lower cooling needs. A more detailed discussion of the energy requirements and heat integration is provided in Section 3.3.

Regarding the products composition, TPA has the targeted 99.5 wt% purity in both layouts with water as main impurity (0.3–0.4 wt%) and other compounds present in ppm levels (bottom left chart). Increasing the AP recirculation to 70% slightly reduces the water content to match the required purity but increases the concentration of dissolved contaminants by a factor of 3–6, with EG and MHET being close to 1000 ppm and BHET, IPA and DEG in the range of 50–200 ppm. Future studies are recommended to test the solubility of the by-products in aqueous solutions containing different concentrations of EG and TPA, as well as experimental product analysis to improve the simulation results and have a better understanding of the characteristics of the recovered TPA in comparison with commercial polymer grade TPA, given the potential impact of impurities on the downstream polymerization process to PET.

Last, the composition of the wastewater (AP to WWTP) in both layouts is shown in the bottom right chart of Fig. 2. Recovering EG and the poorly soluble by-products results in a tenfold reduction in the total organic load, expressed as chemical oxygen demand (COD), compared to the layout when no EG is recovered. Thus, although the overall mass flow of wastewater generated in the two layouts differs only by 3–10%, the organic load is 10 times lower when EG recovery is implemented.

Overall, these results support the implementation of EG recovery at a high AP recirculation up to 70% as a favorable strategy to significantly reduce the make-up water by 68% and lower the electricity and steam requirements for EG recovery by 40 and 80% respectively, at the expense of an 8% increase in natural gas consumption. Thus, the remaining discussion is focused on the results at 70% AP recirculation. It is worth noticing that although 70% is deemed as the most favorable recirculation ratio constrained by the experimentally supported data that there are no changes in reaction chemistry up to this point [10], future studies including reaction kinetics are recommended to evaluate the impacts of compounds accumulation and higher EG concentration on the reaction chemistry and downstream separation in a more rigorous way, further improving the simulation results.

3.2. Overall mass and carbon balances at 70% AP recirculation

Fig. 3-top shows the results of the overall mass balance at 70% aqueous phase recirculation for the two layouts. When both monomers are recovered, the process yields 0.80 kg of TPA and 0.22 kg of EG per kg of PET respectively and due to the 70% AP recirculation requires only 3.1 kg of make-up water per kg of PET for the depolymerization process, with additional 0.8 kg for TPA washing. For the layout without EG recovery, the inputs and TPA yield are the same, but the production of wastewater is higher as it contains all EG and dissolved species separated as liquid and solid residues in the first layout.

In terms of carbon (Fig. 3-bottom), 74% of the carbon is recovered in the TPA product in both layouts. When both monomers are recovered, additional 13.3% is recovered in the EG product increasing the overall carbon recovery to 87.2%, with 13% lost in the solid and liquid residues and wastewater stream. Although valorization of the solid by-product is not evaluated here, this stream contains 7% of carbon in the form of terephthalate monomers (MHET, BHET) whose utilization for direct PET repolymerization has been explored in literature [35,36], being a potentially valuable product in future studies to increase the carbon recovery of the process up to 94.2%. Only 2.7% of the total carbon is lost in the wastewater stream when EG recovery is implemented, versus 26% when no EG is implemented. Fig. 4 presents a more detailed view of the carbon flows within the nHTP process. For reference, the obtained monomer yields of 0.8 kg TPA/kg PET and 0.22 kg EG/kg PET are competitive with existing solvolysis technologies, being slightly lower

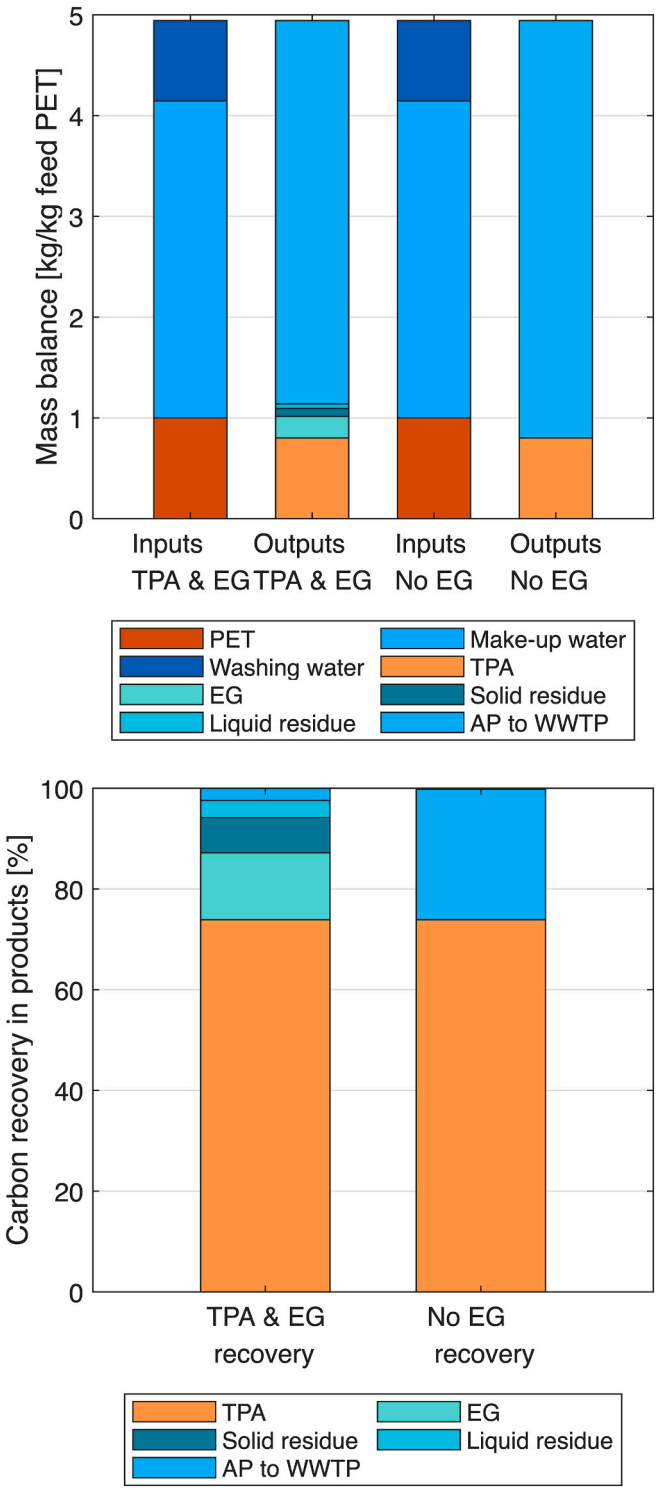

Fig. 3. Overall inputs and outputs (top) and carbon recovery (bottom) of nHTP process layouts at 70% aqueous phase recirculation

than the reported for alkaline hydrolysis [7] (0.84 and 0.34 kg/kg PET for TPA and EG respectively) but higher than for enzymatic hydrolysis [6,31] (0.68 and 0.14 kg/kg PET for TPA and EG respectively).

3.3. Energy requirements at 70% AP recirculation

Fig. 5-top presents an overview of the estimated energy consumption for both layouts at 70% AP recirculation. In the two cases, the largest

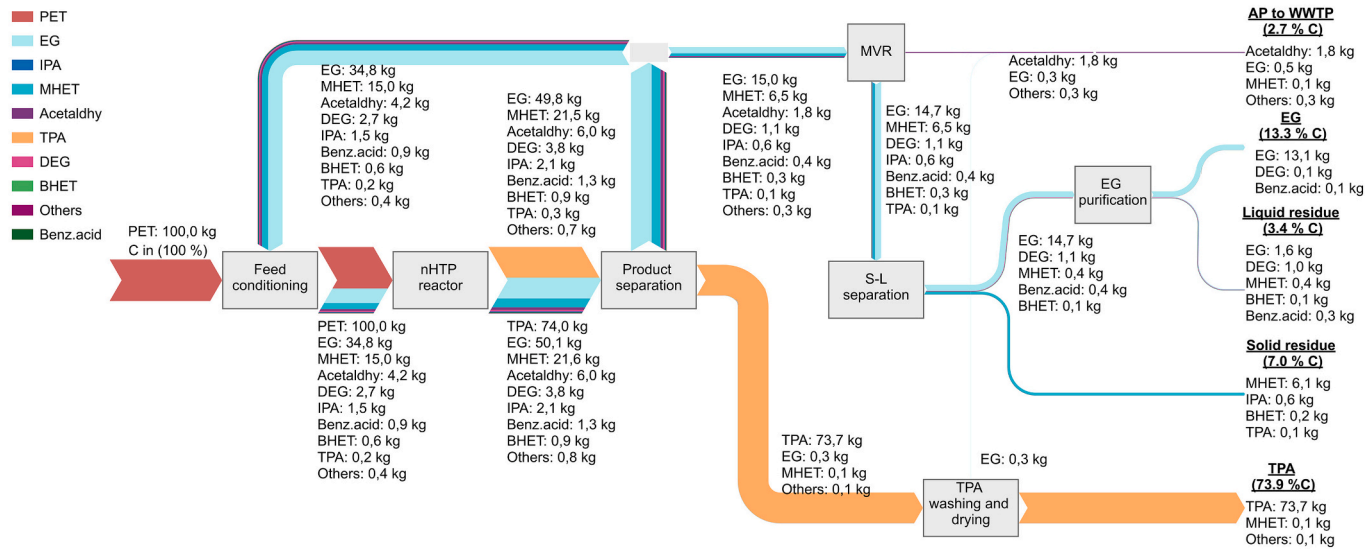

Fig. 4. Carbon distribution across nHTP process streams at 70% aqueous phase recirculation (Relative error across HTP reactor is 0.2% (Table 1).

heating utility is fuel for the furnace to heat up the reactor feed (1.2 kWh/kg PET), followed by a relatively low steam consumption of ~0.25 kWh/kg PET. In the layout where EG is recovered, there is an increased consumption of electricity to run the compressor in the MVR unit (+ 50%) and cooling water for condensers (+ 14%), plus a new demand of high-pressure (HP) steam for reboilers (0.2 kWh/kg PET), which corresponds to unit operations T1, T2 and T3 in Fig. 1. The requirement of low-pressure (LP) steam is the same in the two configurations as it is only used for TPA drying.

Cooling water requirements are substantial (1.5–1.7 kWh/kg PET) and largely attributed to condensation of the aqueous phase prior to pumping and recirculation (V3 unit, 1.2 kWh/kg PET at ~100 °C). This excess heat constitutes a waste for the process in the absence of enough heat requirements below this temperature, as visualized in the grand composite curves in Fig. 5- bottom; however, it holds potential for integration in district heating networks. Although heat integration in the process achieves total hot utilities that are only 6% higher than the minimum estimated from pinch analysis, there is room for improvement in the valorization of excess heat within the process (i.e. heat pumps), or via integration with external processes or district heating networks. It is worth noting that even at 70% AP recirculation the EG concentration remains below 10 wt%, which ultimately makes the EG recovery process highly energy intensive via distillation despite the large savings achieved by the MVR system. Additionally, due to the relatively high fuel consumption, further improvements via intensified approaches such as high temperature heat pumps and integration with other high temperature processes for heat cascade are recommended.

For reference, an overview of the estimated energy requirements and the reported in literature for other hydrolysis processes is shown in Table 5. Since the nHTP process does not require chemicals during the reaction nor the need for regeneration, relatively low electricity and steam consumption can be expected. However, this benefit comes at the expense of higher natural gas consumption to produce the high temperature heat for the depolymerization process, which in alkaline/enzymatic hydrolysis is not required.

3.4. Cost estimation, small vs. large scale

Fig. 6 presents the results of the TPA production cost for the two layouts at small and large scale and 70% AP recirculation. The variation between low and high-cost scenarios is represented by the shadow area with the center line as the baseline cost.

At small scale, the TPA cost is about the same for both layouts at 2.39 EUR/kg in the baseline scenario, varying between 2.1 and 2.7 EUR/kg in the low and high respectively. Although the layout in which EG is recovered has a larger CAPEX and fixed costs, the significant reduction in wastewater treatment costs pays off the investment, being even more beneficial for the overall economics of the process than the EG credits themselves. In terms of price sensitivity, the production cost when EG recovery is implemented is more sensitive to CAPEX than when no EG is recovered (Fig. 6 -right), while in the layout with no EG recovery the impact of CAPEX is attenuated because of the direct influence it has on O&M and fixed costs, estimated as a factor of CAPEX, despite the fact that the PET feed cost has the highest share in the TPA production cost.

Increasing the plant size to 300 t/d reduces the TPA cost by about 50% to 1.18 EUR/kg (1–1.4 EUR/kg) in the layout when both monomers are recovered, and by 35% to 1.56 EUR/kg (1.4–1.8 EUR/kg) in the layout with no EG recovery, having a higher impact in the first case because this layout is more CAPEX intensive. A more detailed look into the CAPEX distribution and the total installed cost at different aqueous phase recirculation is presented in Fig. 7. For the process layout with both TPA and EG recovery, 40% of the total installed equipment cost corresponds to the EG recovery section and about half of it is the mechanical vapor recompression system (MVR).

Although scaling-up makes the TPA cost more competitive with the virgin TPA market price in Europe and USA (0.77–1.03 USD/kg [37]), it is worth noticing that the result is highly sensitive to the price of the PET feedstock. Regardless of the process layout, the PET feed represents about 0.7–0.8 EUR/kg TPA, leaving very little margin for cost reductions in CAPEX or OPEX via process improvements when targeting cost-parity with the virgin material. This evidences the need to explore the utilization of lower-cost feedstock such as mixed waste and textiles and to assess the impact of utilizing more complex feedstock in process performance and production costs. These are key aspects to address in future techno-economic assessments of the nHTP technology.

Regarding cost estimates for competing hydrolysis processes, a similar TPA cost of ~2.5 EUR/kg (2.3 USD/kg) has been reported for alkaline hydrolysis at a scale of 24 t/d PET [38], while for enzymatic hydrolysis the estimates are higher with ~2.7 EUR/kg (2.43 USD/kg) at 50 t/d scale and ~2 EUR/kg (1.84 USD/kg) at 300 t/d [6], indicating

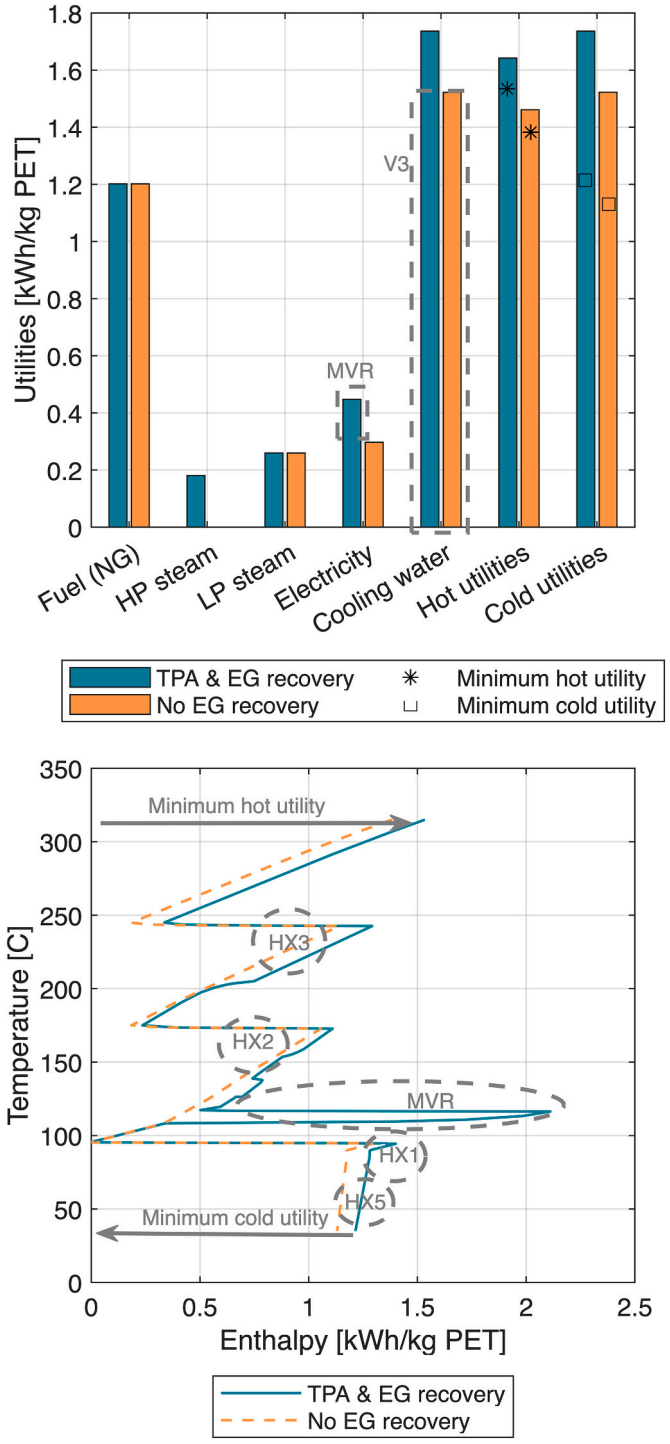

Fig. 5. Specific energy consumption (top) and grand composite curves (bottom) of nHTP process layouts at 70% aqueous phase recirculation

relatively higher costs for this technology compared to the nHTP and alkaline counterparts.

3.5. Carbon footprint of the nHTP process and competing solvolysis technologies

Fig. 8 shows the results of the carbon footprint of the nHTP process and the alkaline and enzymatic hydrolysis for the low and high CO_2 emission scenarios. The mass and energy balances can be consulted in the supplementary document (Section S4).

Table 5
Overview of estimated energy requirements for the nHTP process at 70% recirculation and the reported in literature for alkaline and enzymatic hydrolysis.

kWh/kg PET flake	nHTP – TPA & EG recovery	nHTP – No EG recovery	Alkaline hydrolysis [7]	Enzymatic hydrolysis [31]
Electricity	0.45	0.30	0.98	0.77[a]
Natural gas	1.20	1.20	0.00	0.00
Steam	0.44	0.26	2.26	0.83
Cooling water	1.74	1.52	3.28	3.41

[a] Reported value in source is 1.756 kWh/kg TPA and includes pretreatment of PET bale into flakes. Recalculated value to PET flake basis is based on a yield of 0.63 kg TPA/kg PET flake minus pretreatment electricity from PET bale into flakes of 0.34 kWh/kg PET flake [31].

The results for the nHTP process indicate a lower carbon footprint for the layout when both monomers are recovered. In the low scenario (Fig. 8-left), the carbon footprint of the recovered TPA is 0.98 and 1.67 kg CO_2e/kg TPA in the two layouts, with and without EG recovery respectively, being approximately 30% lower when EG is recovered from the process due to the EG credits. The results obtained for the alkaline and enzymatic hydrolysis in the low scenario are similar (~1–1.3 kg CO_2e/kg TPA), and based on these, nHTP and alkaline hydrolysis show the best performance being ~50% below the virgin TPA. In the high scenario (Fig. 8-right) the trend is the same but the results increase by 0.5–0.6 kg CO_2e/kg TPA in the first three cases, mainly attributed to the PET feedstock and make-up water impacts, and by 1.4 kg CO_2e/kg TPA in the enzymatic hydrolysis process due to higher impacts of chemicals and waste disposal. The enzymatic hydrolysis results are within the reported in referenced LCA studies of 0.9–4.27 kg CO_2e/kg TPA [31].

The largest GHG contributor in the nHTP process for the low and high estimates is the PET feedstock with 52–57% (0.85–1.06 kg CO_2e/kg TPA) which includes collection, sorting and shredding into flakes, followed by the natural gas with 18–23% (0.35 kg CO_2e/kg TPA) and steam with 10–12% (0.18–0.2 kg CO_2e/kg TPA), indicating the potential for heat electrification or low-carbon heat options to achieve around 25–35% GHG emissions reductions. In the alkaline hydrolysis process, the larger contributions from steam and electricity are due to the higher energy consumption for product separation and salt regeneration, while in the enzymatic hydrolysis process, the impacts of chemicals and waste disposal are significant since the process design reported does not include recovery of the acid/base used, negatively affecting its GHG emission performance. Future studies are recommended to evaluate the environmental impacts of the process based on a more comprehensive LCA methodology, including emissions sources not included in the present study such as the embodied carbon emissions related to the infrastructure, as well as other impact categories beyond global warming such as freshwater consumption, ecotoxicity, human toxicity, and acidification among others.

4. Conclusions

This study evaluates the techno-economic feasibility and carbon footprint of PET chemical recycling via neutral hydrolysis (nHTP), using process simulation and experimental data at continuous-lab scale to assess the recovery of terephthalic acid (TPA) and ethylene glycol (EG) monomers in a scaled-up process at 24 t/d and 300 t/d PET feed.

Based on the evaluation of two process layouts, with and without the recovery of EG, it is demonstrated that the implementation of EG recovery is favorable to enhance waste valorization by recovering an additional product and drastically reduce the organic load of the wastewater generated in the process by a factor of 10. This is due to the separation of additional compounds from the wastewater during the EG

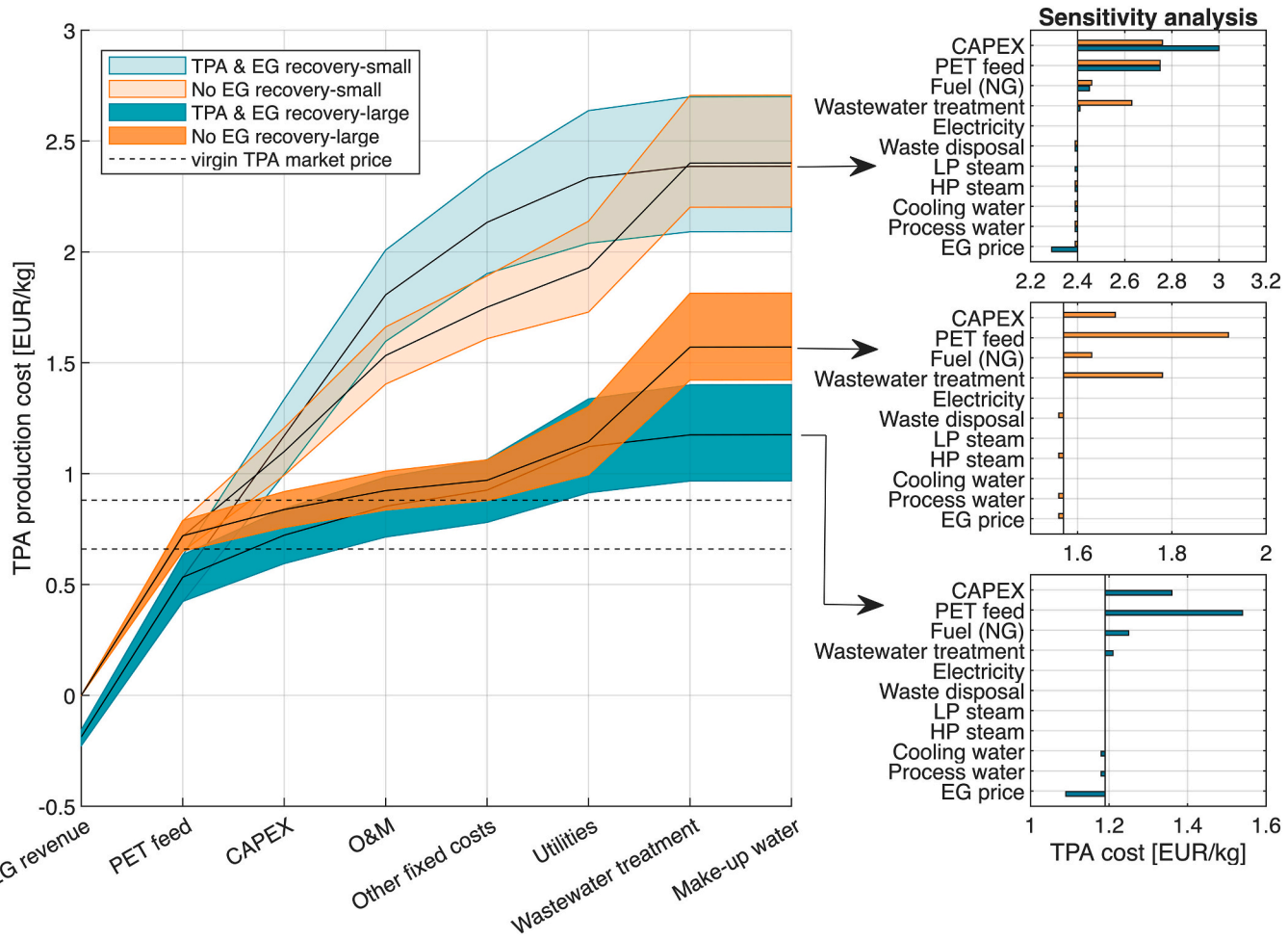

Fig. 6. TPA production cost in nHTP process layouts at 70% aqueous phase recirculation (left) and sensitivity of TPA cost to different cost contributors (right).

recovery process, mainly BHET and MHET that hold valorization potential as PET intermediate. The results support the implementation of a high AP recirculation up to 70% as a favorable strategy to significantly reduce the make-up water consumption by 68% and lower the electricity and steam requirements for EG recovery by 40 and 80% respectively, at the expense of an 8% increase in fuel consumption in the nHTP reactor that is relatively low compared to the estimated savings.

At the highest aqueous phase recirculation of 70%, the nHTP process yields 0.80 kg of TPA and 0.22 kg of EG per kg of PET at a purity ≥99 wt % using 0.45, 0.44 and 1.2 kWh/kg PET of electricity, steam and natural gas respectively. While the use of electricity and steam is 40–80% lower than the reported in literature for competing hydrolysis technologies (e.g. alkaline and enzymatic hydrolysis), fuel is required due to the relatively higher reaction temperatures in the nHTP process which indicates the need to explore sustainable options for high temperature heat in the process.

In terms of costs, implementing the recovery of EG is significantly more beneficial for the economics of the process in terms of reducing the wastewater treatment costs than in providing additional EG revenues. This cost reduction largely compensates the higher capital investment at small scale yielding the same TPA cost in both layouts at 2.4 EUR/kg, while at large scale the TPA cost is 24% lower when EG recovery is implemented with 1.2 EUR/kg. Overall, these results are comparable to the reported in literature for alkaline hydrolysis, and lower than current estimates for enzymatic hydrolysis. Based on the analysis, although scaling-up achieves significant cost reduction, the PET feed (PET flakes) cost is the main contributor with 0.7–0.8 EUR/kg TPA which clearly highlights the need to evaluate the performance of lower-cost feedstocks such as mixed waste and textiles in future assessments.

The results of the carbon footprint show estimated GHG emissions of 0.98–1.67 kg CO_2e/kg TPA recovered for the best layout in which EG is also recovered, approximately 40% lower than the case when EG is not recovered. PET feedstock and natural gas are the main GHG contributors which reinforce the need to explore alternative feedstocks and low-carbon heat sources. Sustainable heat sources could potentially reduce GHG emissions by 25–35%. Although the carbon footprint results are similar for alkaline hydrolysis and lower than for enzymatic hydrolysis, a more comprehensive LCA assessment is needed in the future to evaluate other environmental impacts. Future studies are recommended to assess the utilization of lower-cost feedstocks (mixed waste and textiles), valorization of the solid by-product, and system integration possibilities in a broader chemical recycling context.

CRediT authorship contribution statement

Eliana Lozano Sanchez: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Antonio Jaime-Azuara:** Writing – review & editing, Visualization, Validation, Methodology, Data curation, Conceptualization. **Aiman Shabbir:** Writing – review & editing, Methodology, Data curation. **Ana Somoza Tornos:** Writing – review & editing, Methodology, Conceptualization. **Thomas Helmer Pedersen:** Writing – review & editing, Methodology, Conceptualization.

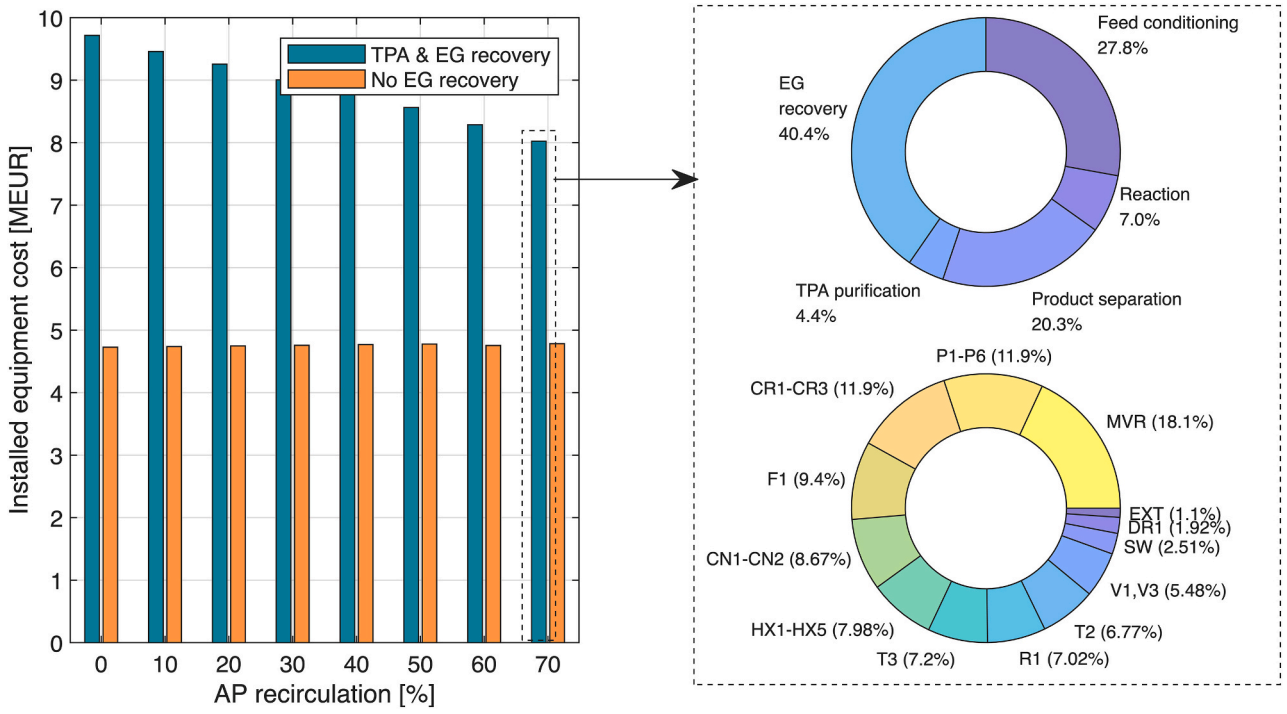

Fig. 7. Total installed cost of nHTP process at increasing aqueous phase recirculation (24 t/d PET feed) (left), and cost breakdown by process section and equipment in layout including TPA and EG recovery (right).

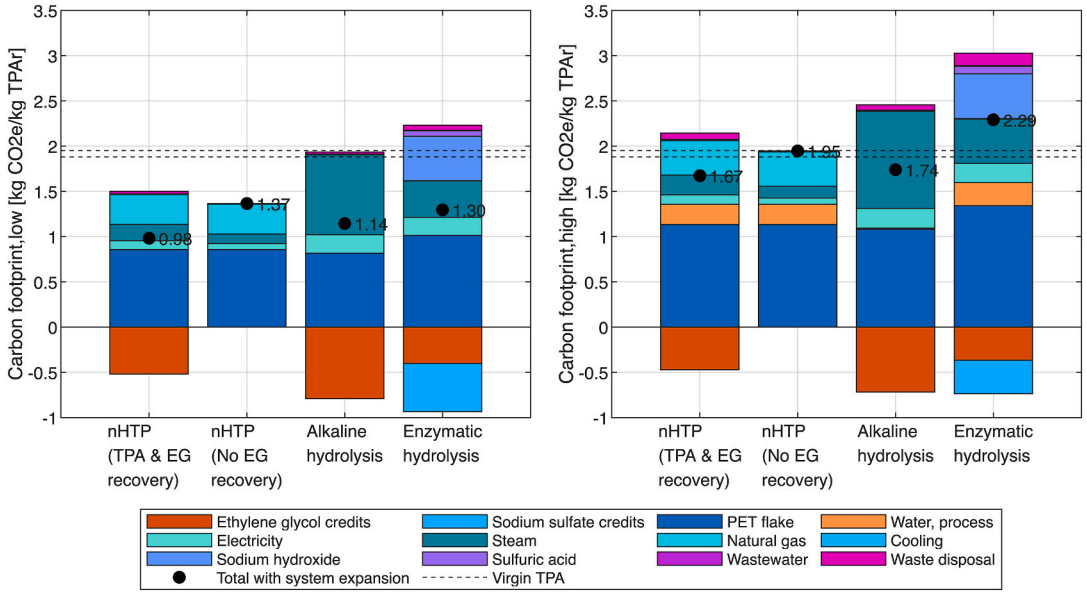

Fig. 8. Carbon footprint results in low (left) and high (right) scenarios for nHTP process and competing solvolysis technologies.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2026.177461.

Data availability

Data will be made available on request.

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